Non-equilibrium quantum condensation in an incoherently pumped dissipative system

M. H. Szymańska,[1] J. Keeling,[2] and P. B. Littlewood[3]

[1] *Clarendon Laboratory, Department of Physics, University of Oxford, Parks Road, Oxford, OX1 3PU, UK*
[2] *Department of Physics, Massachusetts Institute of Technology, 77 Mass. Ave., Cambridge, MA 02139, USA*
[3] *Cavendish Laboratory, University of Cambridge, Madingley Road, Cambridge CB3 0HE, UK*

We study spontaneous quantum coherence in an out of equilibrium system, coupled to multiple baths describing pumping and decay. For a range of parameters describing coupling to, and occupation of the baths, a stable steady-state condensed solution exists. The presence of pumping and decay significantly modifies the spectra of phase fluctuations, leading to correlation functions that differ both from an isolated condensate and from a laser.

PACS numbers: 05.70.Ln, 03.75.Gg, 03.75.Kk, 42.50.Fx

The phenomenon of condensation, i.e macroscopic occupation of a single quantum mode, has attracted considerable attention in recent years. It ranges from Bose-Einstein Condensation (BEC) of structureless bosons to the BCS-type collective state of fermions and has been studied in several physical systems such as degenerate atomic gases and superconductors [1]. Further, recent experimental advances in manipulation of atomic Fermi gases have led to realisation of the BCS-BEC crossover regime [2]. The next challenge is to control and study condensed states in solid-state. For this, the currently promising candidates are excitons in coupled quantum wells [3], microcavity polaritons [4, 5, 6], quantum Hall bilayers [7], and Josephson junction arrays in microwave cavities [8].

Unlike atomic gases, solid-state systems face dephasing and decay, as (with the special exception of superconductors) it is not usually possible to isolate the condensate from the environment: Phonons and impurities lead to dephasing, and due to imperfect trapping, particles escape, requiring external pumping to sustain a steady-state. If such processes are faster than thermalisation the system remains out of thermal equilibrium. Dissipation and decay not only present experimental obstacles, but also pose fundamental questions about the robustness of a condensate: Is a steady-state condensate possible with incoherent pumping and decay, if so, how does it differ from thermal equilibrium? Condensation in dissipative systems also provides a connection to the laser [9]. The relation between lasing and BEC is particularly relevant for polariton BEC, where the experimental distinction between the two is not straightforward [10].

Models that combine potentially strong non-equilibrium pumping with spontaneous symmetry breaking are not well-studied, which motivates a study of simple models to extract the principal qualitative features. In this Letter we study spontaneous condensation in a system coupled to independent baths, not in thermal or chemical equilibria with each other, providing incoherent pumping and decay. We focus on a Bose-Fermi system with disorder localised fermions; this is a model for exciton-polaritons [4, 5, 6] or Josephson junctions in microcavities [8]. However, many of our conclusions apply more generally to condensation with pumping and decay. We show that steady-state spontaneous condensation can occur in such systems, and can be distinct from lasing: The condensate can exist at low densities, far from the inversion required for lasing. We study fluctuations about a steady-state condensate and find that the collective modes are qualitatively altered by the presence of pumping and decay: The low energy phase mode (Goldstone, Bogoliubov mode) becomes diffusive at small momenta. By considering the effect of phase fluctuations, we find the decay of correlations, which at large times and distances differs both from that for a thermal equilibrium condensate and from a laser.

Our Hamiltonian is $\hat{H} = \hat{H}_{sys} + \hat{H}_{sys,bath} + \hat{H}_{bath}$, where,

$$\hat{H}_{sys} = \sum_{\alpha} \varepsilon_{\alpha} \left(b_{\alpha}^{\dagger} b_{\alpha} - a_{\alpha}^{\dagger} a_{\alpha} \right) + \sum_{\mathbf{p}} \omega_{\mathbf{p}} \psi_{\mathbf{p}}^{\dagger} \psi_{\mathbf{p}} + \frac{1}{\sqrt{L^2}} \sum_{\alpha} \sum_{\mathbf{p}} \left(g_{\alpha,\mathbf{p}} \psi_{\mathbf{p}} b_{\alpha}^{\dagger} a_{\alpha} + \text{h.c.} \right), \quad (1)$$

describes two fermionic species b_α and a_α, interacting with bosonic modes $\psi_{\mathbf{p}}$, and coupled to three baths;

$$\hat{H}_{sys,bath} = \sum_{\alpha,k} \Gamma^{a}_{\alpha,k} (a_{\alpha}^{\dagger} A_k + \text{h.c.}) + \Gamma^{b}_{\alpha,k} (b_{\alpha}^{\dagger} B_k + \text{h.c.}) + \sum_{\mathbf{p},k} \zeta_{\mathbf{p},k} (\psi_{\mathbf{p}}^{\dagger} \Psi_k + \text{h.c.}), \quad (2)$$

given by $\hat{H}_{bath} = \sum_k \omega_k^{\Gamma^a} A_k^{\dagger} A_k + \sum_k \omega_k^{\Gamma^b} B_k^{\dagger} B_k + \sum_k \omega_k^{\zeta} \Psi_k^{\dagger} \Psi_k$. A single two level system coupled to multiple baths has been recently studied in the context of the Kondo problem [11]. Condensed solutions of eq. (1) have been studied in the context of atomic Fermi gases [12] and microcavity polaritons [13, 14]. In this Letter we focus on microcavity polaritons, so $b_\alpha^{\dagger}, a_\alpha$ describe an electron and hole within a disorder-localised exciton state of energy ε_α. This can also be viewed as a fermionic representation of a hard-core boson or of a spin. These are dipole coupled to cavity photon modes $\psi_{\mathbf{p}}$, with low $\mathbf{p}$ dispersion, $\omega_{\mathbf{p}} \simeq \omega_0 + \mathbf{p}^2/2m_{\text{ph}}$, where $m_{\text{ph}} = (\hbar/c)(2\pi/w)$ is

the photon mass in a 2D microcavity of width w. Due to the finite reflectivity of the cavity mirrors, photons escape, so the system must be pumped (excitons injected) to sustain a steady-state. Incoherent fermionic pumping and photon decay are described by (2) where A_k, B_k are fermionic annihilation operators for the pump baths, while Ψ_k are bosonic annihilation operators for photon modes outside the cavity.

With pumping and decay, i.e. coupling to baths with different temperatures and/or chemical potentials, the system distribution function can be far from thermal and needs to be obtained self-consistently with the system's spectrum. Therefore we use non-equilibrium field theory in the Keldysh path-integral formulation [15]. Integrating over the bath's degrees of freedom and fermion fields yields an effective action for photons. The dependence of the effective action on the bath properties is parameterised by the functions $\kappa(\omega)$, $\gamma(\omega)$, $F_{A,B}(\omega)$, and $F_\Psi(\omega)$. The cavity decay rate $\kappa(\omega) = \pi\zeta^2(\omega)N^\zeta(\omega)$, where ζ is the coupling of the cavity photons to the bosonic modes in eq. (2) and N^ζ is the density of states of these modes. Similarly $\gamma(\omega) = \pi\Gamma^2(\omega)N^\Gamma(\omega)$ where Γ and N^Γ are respectively coupling to, and the density of states of, the fermionic pumping baths. In this work we assume a flat spectrum for the baths, so κ and γ are frequency independent. Frequency dependence is however present in the bath distribution functions; $F_{A,B}(\omega) = 1-2n^{A,B}(\omega)$, and $F_\Psi(\omega) = 1 + 2n^\Psi(\omega)$, where $n^{A,B}, n^\Psi$ are occupations of the baths.

We proceed by extremising the non-equilibrium effective action with a steady-state, uniform, photon field of the form, $\psi(t) = \psi e^{-i\mu_S t}$, giving a non-equilibrium generalisation of the usual gap equation (e.g. [13, 16]):

$$(\omega_0 - \mu_S - i\kappa)\psi = g\mathrm{Tr}(iG^K_{ba}). \tag{3}$$

Here, $iG^K_{ba}(t,t') = \langle a^\dagger_{cl}(t)b_{cl}(t')\rangle$, is the anomalous Keldysh fermionic Green's function, where $a_{cl} = (a_f + a_b)/\sqrt{2}$ and f, b are the forward and backward branches of the Keldysh time contour [15], and is given by:

$$iG^K_{ba}(\omega) = 2\gamma g\psi\frac{(F_A+F_B)\omega + (F_B-F_A)(\tilde\epsilon + i\gamma)}{[(\omega-E)^2+\gamma^2][(\omega+E)^2+\gamma^2]}, \tag{4}$$

where $E = \sqrt{\tilde\epsilon^2 + g^2|\psi|^2}$, $\tilde\epsilon = \epsilon - \mu_S/2$, and the arguments of $F_{B,A}(\omega)$ are shifted by $\pm\mu_S/2$ while ω is a real frequency. The bosonic bath's distribution F_Ψ does not enter the mean-field gap equation, as the mean-field considers only the condensed photons, and not the population of incoherent photons. In the limit $\gamma, \kappa \to 0$ with the bath distributions $F_{A,B}$ being thermal, eq. (3) reduces to its equilibrium form [13, 14]; for finite γ and κ it is significantly altered.

As in thermal equilibrium, the normal state $\psi = 0$ is always a solution of eq. (3), but for some range of parameters there is also a condensed $\psi \neq 0$ solution. When a solution $\psi \neq 0$ exists, the solution $\psi = 0$ becomes unstable. To understand this instability, we consider small fluctuations about the mean-field, $\psi = \psi_0 + \delta\psi$. The effective action for these fluctuations $\delta\psi$ has a part from the free photon action, and a part from interactions with fermions, $\frac{1}{2}\mathrm{Tr}(G\delta G^{-1}G\delta G^{-1})$, where the fermionic Green's functions G are four by four matrices in the Keldysh and particle-hole (a, b) spaces. Inverting the effective action for fluctuations gives the photon Green's functions, $\mathcal{D}^{K,R,A}$. The retarded and advanced Green's functions $\mathcal{D}^{R,A}$ give the excitation spectrum. The distribution function F_S , defined by $\mathcal{D}^K = \mathcal{D}^R F_S - F_S\mathcal{D}^A$, determines how the spectrum is occupied. As the system need not be in thermal or chemical equilibria with the baths, F_S is in general not thermal, and differs from the bath distributions, $F_{A,B}$ and F_Ψ.

The instability of the normal state when a condensed solutions exists is analogous to that in thermal equilibrium. Even when the bosonic distribution F_S is far from thermal, as the system approaches the phase transition, F_S will have a divergence at a frequency which we define as an effective chemical potential, where $\Im(\mathcal{D}^{R^{-1}}(\mu_{\text{eff}})) = 0$. Taking the zeros of $\Re(\mathcal{D}^{R^{-1}}(\omega^*, q))$ as defining the normal modes of the system, condensation occurs when μ_{eff} reaches the bottom of the band of excitations, as in equilibrium. This condition, that a solution to $\mathcal{D}^{R^{-1}}(\mu_{\text{eff}}, q = 0) = 0$ exists, is equivalent to the gap equation, eq. (3), at $\psi = 0$, $\mu_{\text{eff}} = \mu_S$, since D^R describes a susceptibility which diverges at the transition. Beyond this point, the normal state is unstable, as μ_{eff} would lie in a bosonic band. This idea of instability can be directly connected to another definition: Beyond this point, the poles of the $\mathcal{D}^R$ have positive imaginary parts, fluctuations grow (rather than decay) in time exponentially. To see this, consider the imaginary parts of the poles of $\mathcal{D}^R$ as a function of momentum, q. At large q, these poles describe bare photons, and so are stable. By the previous definitions, above the transition, there is a q at which the normal state Green's function has a real pole, $\mathcal{D}^{R^{-1}}(\omega^*, q) = 0$. The sign of the imaginary part of the pole changes at this point, so the low q poles are unstable. Thus, whenever there is a condensed solution to the gap equation the $\psi = 0$ solution is always unstable.

There is, however, an important difference between the non-equilibrium steady-state and thermal equilibrium. Unlike thermal equilibrium, there is a range of parameters for which neither the normal state nor condensed solutions of the form $\psi(t) = \psi e^{-i\mu_S t}$, are stable. This is not too surprising as systems similar to (1) are known to follow a complicated or even chaotic dynamics [17]. Although such anharmonic solutions would be of a great interest, we focus here on steady-state condensed solutions of the usual form.

Having understood the stability of solutions, we next solve the gap equation (3) to find the non-equilibrium phase diagram. Since eq. (3) is complex it gives two

equations for two unknowns: the order parameter ψ and the frequency μ_S. The common oscillation frequency μ_S would in thermal equilibrium be the system's chemical potential, considered as a control parameter, adjusted to match the required density, and the (real) gap equation determines only ψ. Here, because different baths have different chemical potentials, the system is not in chemical equilibrium with either bath, so both μ_S and ψ must be found from the gap equation. The density, which can be found given ψ and μ_S, is set by the relative strength of the pump and decay.

From here, the bath distributions are taken to be individually in thermal equilibrium, although the formalism would allow any distribution. However, as the baths need not be in equilibrium with each other, the system can still be far from thermal equilibrium. Further, for simplicity, the figures shown are all for the baths at zero temperature, so the bath distributions are defined by their chemical potentials. To ensure that on average only one of the two fermionic levels a, b is occupied, the chemical potentials of baths A, B are related by $\mu_A = -\mu_B$. Since the cavity photon modes start at energies much above the zero for bulk photon modes, we take the chemical potential of the decay bath to be large and negative. With these restrictions, the baths are described by three parameters, the couplings γ, κ and μ_B parameterising the occupation of the pumping baths.

Figure 1 shows a phase diagram in terms of the parameters γ, κ, μ_B. It shows both where a condensed solution to the gap equation exists, and where the solution is stable. At a given decay rate κ there is a minimum γ (as pumping is proportional to γ) and a maximum γ (as dephasing is also proportional to γ) required for condensation. Stable condensed solutions exist only for κ smaller than about $0.2g$. In the region shown in Fig. 1 the condensed solutions are all below population inversion. However for $\gamma > g$, when system is in a weak coupling regime, only laser-like solutions which require population inversion are possible. For large γ and μ_B (large pumping), our theory recovers the regular laser limit.

We now discuss the collective modes, considering fluctuations about the steady-state. Motivated by microcavity polaritons, we study these collective modes by calculating the photoluminescence (PL) spectra: $i\mathcal{D}^{<}_{\psi^\dagger\psi}(t,t') = \langle \psi^\dagger_f(t)\psi_b(t')\rangle$. In the normal state, as expected, one finds homogeneously broadened upper and lower polariton modes. This broadening depends on all of the bath parameters, κ, γ and μ_B. Approaching the phase boundary from the normal side, the lower polariton linewidth reduces to zero. This can be understood by identifying the zeros of the real part of the inverse Green's function as the polariton energies, $\Re(\mathcal{D}^{R\,-1}(\omega^*, q)) = 0$, and the imaginary part as giving the linewidth, $1/\tau_p \approx \Im(\mathcal{D}^{R\,-1}(\omega^*, q))$. The earlier discussion relating the gap equation to $\mathcal{D}^{R\,-1}(\mu_S, q = 0)$ implies that $1/\tau_p$ vanishes at the transition.

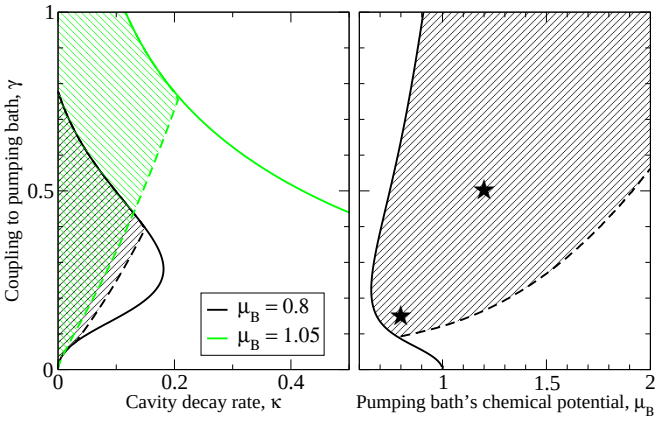

FIG. 1: Mean field phase diagrams. Left: Critical γ vs decay rate κ, for two different pumping bath chemical potentials, μ_B. Right: Critical γ vs μ_B for κ=0.038g as in experiment [6]. Solid lines show where a solution of the gap equation exists, dashed lines show where that solution is stable. The region where a stable condensed solution exists is shaded. Stars mark the choice of parameters shown in Fig. 2.

In the condensed state, as in equilibrium[13], the excitation spectrum changes. This leads to a soft mode, describing phase fluctuations, as global phase rotation symmetry is broken. These phase fluctuations may be large, so one must include the phase fluctuations to all orders [16] to find the field-field correlations; amplitude fluctuations remain small, as they have a restoring force. Writing, $\psi(t) = \sqrt{\rho_0 + \pi(t)}e^{i\phi(t)}$, where ρ_0 is the mean-field condensate density, the PL spectrum is thus:

$$i\mathcal{D}^{<}_{\psi^\dagger\psi}(t,r) = \rho_0\left[1 + \mathcal{O}\left(1/\rho_0\right)\right]\exp\left[-f(t,r)\right], \quad (5)$$

$$f(t,r) = \int d\nu \int (dq)^2 \left[1 - e^{i(\nu t + \mathbf{q}\cdot\mathbf{r})}\right] i\mathcal{D}^{<}_{\phi\phi}(\nu, q). \quad (6)$$

The phase-phase Green's function $\mathcal{D}^{<}_{\phi\phi}$ is found by inverting the action expanded to quadratic order.

The $\mathcal{O}\left(1/\rho_0\right)$ terms in eq. (6), due to amplitude-amplitude and phase-amplitude Green's functions, are included when plotting Fig. 2. The form of these terms, and the full expression for $\mathcal{D}^{<}_{\phi\phi}$, are complex, so we do not reproduce them here. The behaviour of $\mathcal{D}^{<}_{\phi\phi}$ in the limit of small frequencies and momenta, is of interest, and takes a simple form:

$$i\mathcal{D}^{<}_{\phi\phi}(\nu, q) \approx C\left[\left(c^2q^2 - \nu^2\right)^2 + 4\nu^2x^2\right]^{-1}, \quad (7)$$

where C, c, and x can be found from the full expressions. Without pumping and decay, the term x would vanish. Its presence means that rather than a linear dispersion at low q, as in a closed system in thermal equilibrium[13, 16], the poles of the Green's function have the following form $\nu = -ix \pm i\sqrt{x^2 - c^2q^2}$. For $|q| < x/c$, these modes

are diffusive; the poles are entirely imaginary, only for $|q| > x/c$, do they acquire a real part. This behaviour is apparent in the PL spectrum in Fig. 2. The diffusive behaviour at small q is not limited to Bose-Fermi systems; it should also be present in other condensed systems with pumping and decay. Note that at $q = 0$ and $\nu = 0$, $\mathcal{D}^{<}_{\phi\phi}$ has a real pole; a manifestation of broken symmetry in the infinite system.

As given by eq. (5), the PL spectrum does not depend linearly on $\mathcal{D}^{<}_{\phi\phi}$. If $\mathcal{D}^{<}_{\phi\phi}$ were small, the exponential in eq. (5) could be expanded, giving PL divided between a condensate term $\rho_0\delta(\nu)\delta(q)$, and a part from occupation of the fluctuation spectrum. However, at $\nu, q \to 0$, $\mathcal{D}_{\phi\phi}$ is not small, and so phase fluctuations give a lineshape to the condensate and determine long time field-field correlations. In 2D, inserting eq. (7) in eq. (6), the main dependence of $f(t,r)$ on t, r comes from a logarithmic divergence, $\int dq/q$, cut at large momenta by terms beyond those in eq. (7), and at small momenta by one of $1/r, 1/ct, 1/c\sqrt{t\tau_g}$ or $1/c\tau_g$, where $\tau_g = 1/x$ is the lifetime of the phase mode. This logarithmic form leads to power law field-field correlations. (This is true also in thermal-equilibrium[16], for which, $f(t,r) = \eta \ln\left(\sqrt{c^2t^2 + r^2}/\beta c\right)$, with $\eta \propto k_BT/\rho_0$.) According to the relative values of r, t, τ_g, different lower cutoffs apply, and so the power law differs at different places in the r, t plane. For small r, t we recover the equilibrium power laws; however when $t \gg \tau_g$, and $c\sqrt{t\tau_g} \gg r$, then τ_g becomes important, and $f(t) \propto \ln(c\sqrt{t\tau_g})$, giving a condensate lineshape that differs from that of an non-dissipative, thermal equilibrium 2D system. Power law field correlations at long times lead to a power law divergence of their Fourier transform as $\nu, q \to 0$, so there is no well defined linewidth. This differs from phase diffusion for a single mode studied in Laser theory[9], where phase correlations grow linearly in time, giving exponential decay of field correlations.

At higher q and ν (Fig 2 Insets) the difference between expanding to linear order in Green's functions and keeping all orders of phase fluctuations is not visible. The strong coupling spectrum consists of the usual phase and amplitude modes (marked a,b) emerging from the lower and upper polariton branches. One can also see an occupation edge (c), and above that the lower polariton following the exciton dispersion (d). These features at higher momenta are analogous to thermal equilibrium non-dissipative polariton condensation in strong coupling regime [13] but with non-thermal occupations. Increasing pumping, dephasing increases, and the system crosses to weak-coupling (lower panels of Fig. 2), the polariton splitting is suppressed, and the spectrum follows the photon dispersion. The diffusive region, then linear dispersion, at small momentum, seen in the main figures, occurs both in strong and weak coupling and is a sign of condensation in the dissipative system.

To conclude, we have studied how steady-state spontaneous condensation emerges in non-equilibrium systems with pumping and decay. This condensation is distinct from lasing: It can occur at densities much lower than the population inversion required for lasing, and the decay of correlations, and thus condensate lineshape, differ from that for phase diffusion of a single laser mode. Dissipation qualitatively changes the fluctuation spectrum with respect to isolated thermal equilibrium: At low momenta, the phase mode becomes diffusive, changing the power-laws controlling long time decay of field-field correlations. These conclusions, although studied here for microcavity polaritons, apply also to other Bose and Bose-Fermi condensates subject to pumping and decay.

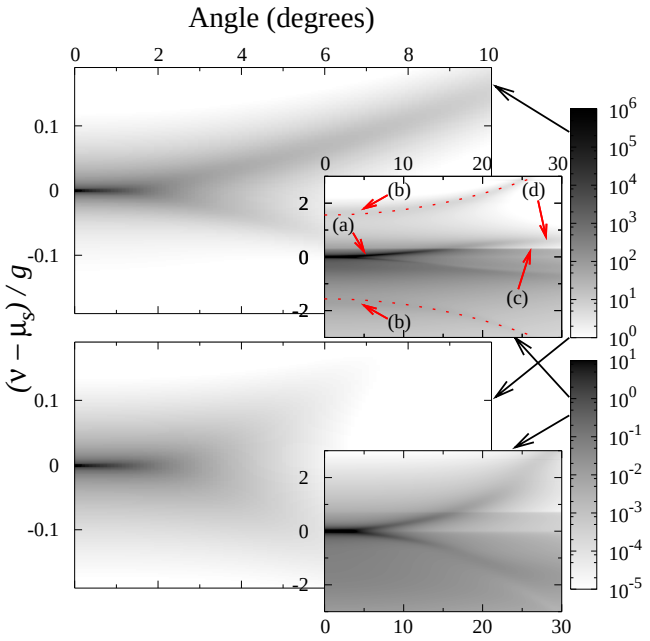

FIG. 2: Photoluminescence $i\mathcal{D}^{<}_{\psi^\dagger\psi}(\nu, q)$ of a condensed system, where q is shown by angle of emission $\tan^{-1}(cq/\omega_0)$. Top: Strong coupling, Bottom: weak coupling, exact parameters marked by stars on Fig. 1. Main figures show PL from small ν, q region, to all orders in phase fluctuations. Insets show a larger range of ν, q for the same parameters. (Dotted lines have been added to show the faint amplitude mode).

We are grateful to B. D. Simons and R. Zimmermann for stimulating discussions. We acknowledge financial support from EPSRC (MHS) and the Lindemann Trust (J.K).

[1] A. Griffin, D. W. Snoke, and S. Stringari, eds., *Bose-Einstein Condensation*, (Cambridge University Press, Cambridge, 1995).

[2] C. A. Regal, M. Greiner, and D. S. Jin, Phys. Rev. Lett. **92**, 040403 (2004); M. W. Zwierlein *et al.*, Phys. Rev. Lett. **92**, 120403 (2004).

[3] L. V. Butov *et al.*, *Nature*, **417**, 47 (2002); **418**, 751 (2002); D. Snoke *et al.*, *Nature*, **418**, 754 (2002).
[4] Le Si Dang *et al.*, Phys. Rev. Lett. **81**, 3920 (1998).
[5] H. Deng *et al.*, Science **298**, 199 (2002); PNAS **100**, 15318 (2003).
[6] M. Richard *et al.*, Phys. Rev. Lett. **94**, 187401 (2005).
[7] J. P. Eisenstein, A. H. MacDonald *Nature* **432**, 691 (2004).
[8] P. Barbara *et al.*, Phys. Rev. Lett. **82**, 1963 (1999).
[9] H. Haken, in *Quantum Optics*, edited by Kay and Maitland (academic Press, London, 1970), p. 201.
[10] M. H. Szymanska and P.B. Littlewood, Sol. Stat. Comm. **124**, 103 (2002); M. H. Szymanska, P.B. Littlewood, and B. D. Simons Phys. Rev. A **68**, 013818 (2003).
[11] J. Paaske *et al.*, Phys. Rev. B **70**, 155301 (2004)
[12] M. Holland, *et al.*, Phys. Rev. Lett. **87**, 120406 (2001); E. Timmermans, *et al.*, Phys. Lett. A **285**, 228 (2001); Y. Ohashi, A. Griffin, Phys. Rev. Lett. **89**, 130402 (2002).
[13] J. Keeling, *et al.*, Phys. Rev. Lett. **93**, 226403 (2004); Phys. Rev. B **72**, 115320 (2005).
[14] F. M. Marchetti *et al.*, Phys. Rev. Lett. **96**, 066405 (2006).
[15] A. Kamenev, in *Nanophysics: Coherence and Transport* (Elsevier, 2005), p 177.
[16] N. Nagaosa, *Quantum field theory in strongly correlated electronic systems* (Springer-Verlag, 1999).
[17] C. Emary, T. Brandes, Phys. Rev. Lett. **90**, 044101 (2003).